PASSION FOR PRECISION

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by

Theodor W. Hänsch

Max-Planck-Institut für Quantenoptik, Garching, and
Department of Physics, Ludwig-Maximilians-Universität München, Germany.

ABSTRACT

Optical frequency combs from mode-locked femtosecond lasers have revolutionized the art of counting the frequency of light. They can link optical and microwave frequencies in a single step, and they provide the long missing clockwork for optical atomic clocks. By extending the limits of time and frequency metrology, they enable new tests of fundamental physics laws. Precise comparisons of optical resonance frequencies of atomic hydrogen and other atoms with the microwave frequency of a cesium atomic clock are establishing sensitive limits for possible slow variations of fundamental constants. Optical high harmonic generation is extending frequency comb techniques into the extreme ultraviolet, opening a new spectral territory to precision laser spectroscopy. Frequency comb techniques are also providing a key to attosecond science by offering control of the electric field of ultrafast laser pulses. In our laboratories at Stanford and Garching, the development of new instruments and techniques for precision laser spectroscopy has long been motivated by the goal of ever higher resolution and measurement accuracy in optical spectroscopy of the simple hydrogen atom which permits unique confrontations between experiment and fundamental theory. This lecture recounts these adventures and the evolution of laser frequency comb techniques from my personal perspective.

INTRODUCTION

In our highly complex and ever changing world it is reassuring to know that certain physical quantities can be measured and predicted with very high precision. Precision measurements have always appealed to me as one of the most beautiful aspects of physics. With better measuring tools, one can look where no one has looked before. More than once, seemingly minute differences between measurement and theory have led to major advances in fundamental knowledge. The birth of modern science itself is intimately linked to the art of accurate measurements.

Since Galileo Galilei and Christiaan Huygens invented the pendulum clock, time and frequency have been the quantities that we can measure with

the highest precision. Today, it is often a good strategy to transform other quantities such as length or voltage into a frequency in order to make accurate measurement. This is what my friend and mentor Arthur Schawlow at Stanford University had in mind when he advised his students: "Never measure anything but frequency!" Measuring a frequency, i.e. counting the number of cycles during a given time interval, is intrinsically a digital procedure that is immune to many sources of noise. Electronic counters that work up to microwave frequencies have long been available. In 1967, the Conference Generale des Poids et Mesures (CGPM) has defined the second, our unit of time, as the period during which a cesium-133 atom oscillates 9 192 631 770 times on a hyperfine clock transition in the atomic ground state. Today, after 50 years of continuous refinement, microwave cesium atomic clocks reach a precision of 15 decimal digits [1].

Even much higher precision is expected from future optical atomic clocks which use atoms or ions oscillating at the frequency of light as the "pendulum". By slicing time into a hundred thousand times finer intervals, such clocks will greatly extend the frontiers of time and frequency metrology. The long missing clockwork mechanism can now be realized with a femtosecond laser frequency comb, an ultra-precise measuring tool that can link and compare optical frequencies and microwave frequencies phase coherently in a single step. Laser frequency combs provide powerful tools for new tests of fundamental physics laws. Precise comparisons of optical resonance frequencies of atomic hydrogen and other atoms with the microwave frequency of a cesium atomic clock are already establishing sensitive limits for possible slow variations of fundamental constants. Optical high harmonic generation is extending frequency comb techniques into the extreme ultraviolet, opening a new spectral territory to precision laser spectroscopy. Frequency comb techniques are also providing a key to attosecond science by offering control of the electric field of ultrafast laser pulses.

Femtosecond laser frequency combs have been highlighted in the citation for the 2005 Nobel Prize in Physics. Although perfected only about seven years ago, they have already become standard tools for precision spectroscopy and optical frequency metrology in laboratories around the world. Commercial instruments have quickly moved to the market, and extensive review articles and books have been written on frequency comb techniques [2 - 4]. In this lecture I will try to give my personal perspective on the evolution of these intriguing measuring tools for time and frequency. Far from attempting a comprehensive review, I have selected references that helped guide my own insights along a winding path.

THE DAWN OF DOPPLER-FREE LASER SPECTROSCOPY

High resolution laser spectroscopy and precise spectroscopic measurements have appealed to me since I was a graduate student at the University of Heidelberg. For my diploma and thesis research I worked with helium-neon gas lasers in the group of Peter Toschek at the Institute of Applied Physics,

headed by Christoph Schmelzer. I was intrigued by the central narrow Lamb dip that Abraham Szöke and Ali Javan had first observed while scanning the frequency of a single-mode gas laser across the Doppler-broadened gain profile [5]. Such a dip had been predicted by Willis Lamb in his semiclassical laser theory [6]. Bill Bennett was the first to give a simple explanation in terms of saturation and spectral hole burning the two counter propagating waves inside the standing wave laser cavity [7]. Other researchers such as John Hall, Veniamin Chebotaev, or Christian Bordé soon explored "inverted Lamb dips" by placing some absorbing molecular gas inside the laser cavity [8]. With resonances of unprecedented spectral resolution, one could almost smell the revolution in laser spectroscopy that would unfold within the next few years. At that time, however, such Doppler free spectroscopy remained limited to the study of gas laser transitions or of a few molecular absorption lines in accidental coincidence. In my own work with Peter Toschek, I studied quantum interference effects in coupled atomic three-level systems [9, 10], demonstrating phenomena which have recently been recognized as important, such as lasing without inversion or electromagnetically induced transparency. They are also essential to understand slow light.

In 1970, I joined Arthur L. Schawlow at Stanford University as a postdoc. Collaborating in separate experiments with Peter Smith, then at Berkeley [11], and with Marc Levenson at Stanford [12], I perfected a new method of Doppler-free saturation spectroscopy that did not require the sample to be placed inside a laser cavity. Soon afterwards, I succeeded in making a nitrogen-laser-pumped widely tunable pulsed dye laser so highly monochromatic that we could apply Doppler-free saturation spectroscopy to arbitrarily chosen atomic resonance lines [13, 14]. Broadly tunable laser action in liquid solutions of organic dyes had been discovered in 1966, independently by Fritz Schäfer [15] and Peter Sorokin [16].

LASER SPECTROSCOPY OF ATOMIC HYDROGEN

Arthur Schawlow at Stanford suggested to apply our technique to the red Balmer-α line of atomic hydrogen that had been at the center of attention of atomic spectroscopists in the 1930s because of suspected discrepancies between the observed line profile and the predictions of Dirac's relativistic quantum theory [17]. In those days, spectroscopists could only observe a blend of unresolved fine structure components because Doppler broadening is particularly large for the light hydrogen atoms. Spectroscopy of the simple hydrogen atom has long played a central role in the history of atomic physics. The visible Balmer spectrum was the Rosetta stone that allowed us to decipher the laws of quantum physics. It has inspired the path breaking discoveries of Niels Bohr, Arnold Sommerfeld, Louis De Broglie, Erwin Schrödinger, Paul Dirac, and even Willis Lamb at the origin of modern quantum electrodynamics.

In 1972, graduate student Issa Shahin and myself were proud to present to Arthur Schawlow a Doppler-free saturation spectrum of the red hydrogen

Balmer-α line, recorded with our pulsed tunable dye laser [18]. The 2S Lamb shift, i.e. the splitting between the $2S_{1/2}$ and $2P_{1/2}$ states that should be degenerate according to the Dirac theory, appeared clearly resolved in the optical spectrum. This was the beginning of a long adventure in precision spectroscopy of the simple hydrogen atom, which permits unique confrontations between experiment and theory. This quest continues today. It has inspired many advances in spectroscopic techniques, including the first proposal for laser cooling of atomic gases [19], and, most recently, the femtosecond laser frequency comb.

Fig. 1 illustrates how the accuracy of optical spectroscopy of atomic hydrogen has advanced over time [20]. Classical spectroscopists remained limited to about six or seven digits of precision by the large Doppler broadening of hydrogen spectral lines. In 1971, our group at Stanford overcame this barrier by nonlinear spectroscopy with a tunable dye laser. Other groups, notably in New Haven, Oxford, and Paris, soon joined in to improve the accuracy by three orders of magnitude over the next two decades. Around 1990, a new barrier appeared: the limits of optical wavelength metrology due to unavoidable geometric wave front errors. Progress beyond a few parts in 10^{10} has been achieved only because we have learned increasingly well how to measure the frequency of light rather than its wavelength. In 2003, the accuracy has reached 1.4 parts in 10^{14} [21]. Further progress is becoming difficult, because we are again approaching a barrier: the limits of how well we know our unit of time, the second. Cesium atomic clocks have been continually refined over the past 50 years [1], as shown by the dashed line in Fig. 1, but the potential for further improvements seems almost exhausted. However, our optical frequency counting techniques make it now feasible to develop optical atomic clocks, based on sharp optical resonances in laser-cooled trapped ions, neutral atoms or molecules. With such clocks future spectroscopic measurements may reach accuracies of parts in 10^{18} and beyond.

In atomic hydrogen, the highest resolution can be achieved on the ultraviolet 1S-2S two-photon resonance with a natural line width of only 1 Hz. First order Doppler shifts cancel if this transition is excited with two counterpropagating laser waves, as was first pointed out by Veniamin Chebotaev [22]. The first Doppler-free spectra have been recorded in our laboratory at Stanford in 1975 [23]. At Garching, we observe this resonance by collinear excitation of a cold hydrogen atomic beam [21]. Starting in 1986, many generations of graduate students and postdocs have made important contributions to advance the state of the art.

Today, the hydrogen atoms are produced by microwave dissociation of molecules and cooled to a temperature of about 6 K by collisions with the walls of a nozzle mounted to a helium cryostat. A collinear standing wave field at 243 nm for Doppler-free two-photon excitation is produced by coupling the frequency-doubled output of a dye laser into a buildup cavity inside the vacuum chamber. Atoms excited to the 2S metastable state after traveling along a path of about 10 cm are detected by applying a quenching electric field and counting the emitted vacuum ultraviolet Lyman-α photons. The

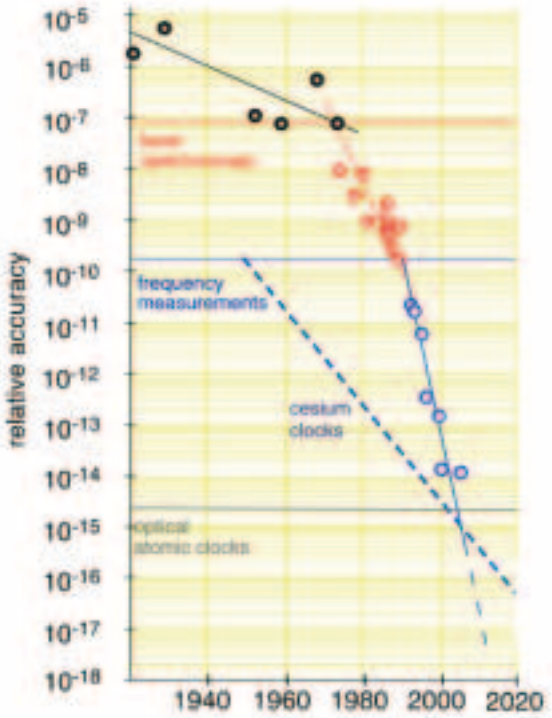

Figure 1. The relative accuracy in optical spectroscopy of atomic hydrogen is charted over eight decades. Major barriers have been overcome in the early 1970s with the advent of Doppler-free laser spectroscopy and in the early 1990s with the introduction of optical frequency measurements. The accuracy of such measurement will soon be limited by the performance of cesium atomic clocks. Dramatic future advances are expected from the development optical atomic clocks.

laser light is periodically blocked by a chopper, and the photon counts are sorted into bins corresponding to different delay times. With slow atoms selected by a delay time of 1.3 ms, the line width is now reduced to about 530 Hz at 243 nm corresponding to a resolution of 4.3 parts in 10^{13}. We would have to reach an accuracy of 5 parts in 10^{15} in order to measure the line position to 1% of this width.

MEASURING OPTICAL FREQUENCIES

The observation of sharp optical resonances by nonlinear laser spectroscopy with a resolution much beyond the measurement limits of wavelength interferometry had long created a strong need for methods to measure the frequency rather than the wavelength of light. The quest for an optical frequency counter is almost as old as the laser itself. Ali Javan, the co-inventor of the helium-neon laser, was the first to superimpose the beams from two different lasers with a beam splitter on a photo detector to observe a beat note, similar to the interference of the sound waves from two tuning forks [24]. This was

an extraordinary result, because it proved that laser light can behave like a classical radio wave. A coherent laser wave can have a well defined phase and amplitude, so that it must be possible to count the ripples of such a light wave. However, at a frequency near 500 000 billion oscillations per second, there are no electronic detectors and circuits fast enough to build an optical frequency counter.

At MIT in the early 1960s, Ali Javan started a research project, aimed at extending microwave frequency counting techniques into the optical spectral region. He experimented with whisker-like metal-insulator-metal point contacts as antennas, detectors and mixers for infrared laser waves. Such elements were later used by John Hall and Ken Evenson at NBS (now NIST) in Boulder to realize the first harmonic laser frequency chain, that was used to determine the speed of light by measuring both the wavelength and the frequency of a methane-stabilized 3.39 μm helium-neon gas lasers [25]. Harmonic laser frequency chains were highly complex systems, engineered to measure just one particular optical frequency, and only a handful of these chains have ever been constructed at a number of well-equipped national metrology laboratories. In the early 1980s, a chain at NBS in Boulder had been perfected so that it could measure the frequencies of some iodine-stabilized visible helium-neon lasers to 10 decimal digits. This demonstration led the Conference Generale des Poids et Mesures in 1983, to redefine the meter by defining the speed of light in vacuum c as exactly 299 792 458 meters per second. One meter is then the distance traveled by light during the time of 1/299 792 458 seconds. From now on, one could determine the precise wavelength of a laser in vacuum, λ, by simply measuring the frequency f, since $f \cdot \lambda = c$.

Unfortunately, the complex NBS frequency chain had to be abandoned soon after this definition was in the books, and for the next decade there was not a single laboratory in the U.S. that could have followed this prescription. A number of European laboratories did better, notably the Observatoire in Paris (now BNM SYRTE) and the Physikalisch-Technische Bundesanstalt (PTB) in Braunschweig. In an article published in early 1996 [26], a team from the PTB laid claim to the first phase coherent frequency measurement of visible radiation. An elaborate frequency chain filling three large laboratories spread over two separate buildings was assembled to compare the frequency of the red intercombination line of atomic calcium with the microwave frequency of a cesium atomic clock. To reach sufficient phase stability, the clock frequency was first reduced to the 100 MHz of a stable quartz oscillator. From here, the chain traversed the entire electromagnetic spectrum in discrete steps, always generating some harmonic frequency in a suitable nonlinear element and producing enough power for the next step with a phase-locked transfer oscillator. A tricky puzzle had to be solved to reach the desired final frequency with the help of several auxiliary oscillators.

It was obvious that we could not afford to assemble such a harmonic laser frequency chain for our hydrogen experiments at Garching. As a simpler alternative, I proposed a frequency interval divider chain in 1988, that worked with

frequency differences rather than the frequencies themselves so that one could stay in a convenient region of the electromagnetic spectrum such as the near infrared where compact diode laser sources are available [27]. The basic building block is an interval divider stage with a laser that is servo-controlled to oscillate at the precise mid-point of two input frequencies. To this end, the second harmonic frequency of the central laser is compared to the sum of the two input frequencies, as generated in a nonlinear optical crystal. With a cascaded chain of n such interval dividers, a large frequency interval can be divided by 2^n. To measure an absolute laser frequency f, one could start with the interval between f and its second harmonic 2f, which is just equal to the frequency f. After repeatedly cutting this interval in half with perhaps 15 stages, the remaining frequency gap is small enough that it can be observed as a beat note with a fast photo-detector and measured with a microwave frequency counter. With Harald Telle, who had joined us from the PTB, and Dieter Meschede, we demonstrated the first working interval divider in 1990 [28].

We never assembled a complete optical frequency counter, but we constructed a chain of four interval dividers to measure a frequency interval of 1 THz that we encountered when comparing our hydrogen 1S-2S frequency with the infrared frequency of a methane-stabilized infrared helium-neon laser at 3.39 μm as the starting point of our own short harmonic laser frequency chain [29]. This intermediate frequency reference had to be repeatedly shuttled to Braunschweig to be calibrated with the PTB frequency chain against a cesium clock. In 1997, we established a new record in optical frequency metrology [29] by determining the ultraviolet 1S-2S frequency to within 3.7 parts in 10^{13}. From this and other spectroscopic measurement in hydrogen, we were able to derive a new value of the Rydberg constant, the scaling factor for any spectroscopic transition, and the most precisely known of the fundamental constants. We could also derive the Lamb shift of the 1S ground state accurately enough to provide a stringent new test of bound-state QED. Assuming that QED is correct we could, moreover, determine new values for the rms charge radius of the proton and the structure radius of the deuteron [29, 30]. We were rather proud that the accuracy achieved with our table-top experiments exceeded that of electron scattering experiments with large accelerators by an order of magnitude.

Soon, a number of metrology laboratories set out to build optical frequency counters based on optical interval division. At Garching, we were also experimenting with electro-optic frequency comb generators kindly provided by Motonobu Kourogi, that could produce an evenly spaced comb of modulation sidebands extending over several THz [31]. A frequency counter could have been realized with only six or seven interval divider stages, if the final frequency gap was bridged with such an electro-optic comb generator. During an extended visit to Garching, Motonobu Kourogi showed us how to observe even feeble comb lines by heterodyne detection, improving the signal to noise ratio with optical balanced receivers and variable beam splitters. We soon verified the accuracy of his frequency comb generator and our frequency interval divider chain in a direct comparison [32].

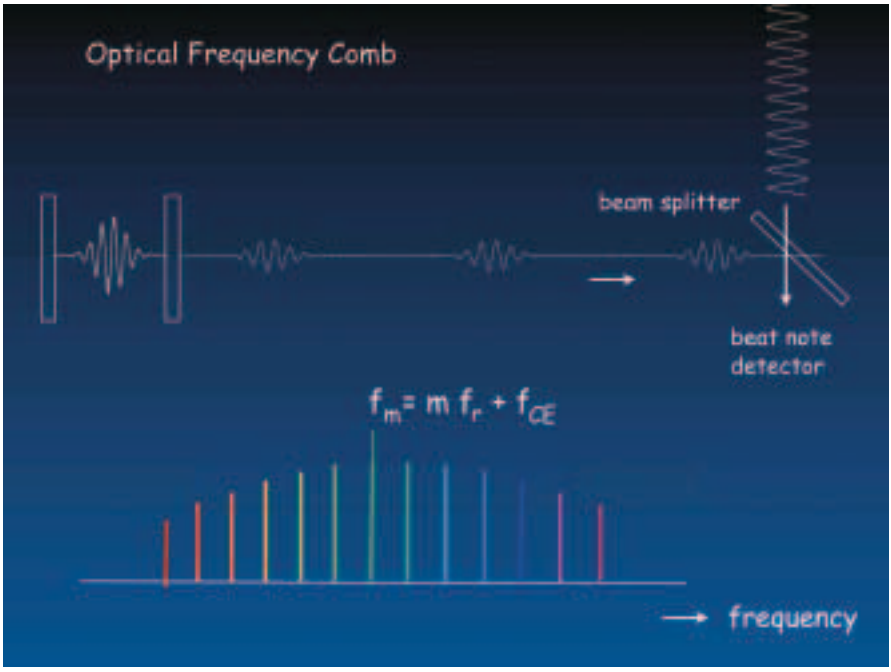

Figure 2. Scheme of a femtosecond laser frequency comb synthesizer.

Many other alternatives have been explored in the long quest for precise optical frequency measurements, including interferometry with modulated laser waves [33, 34] or frequency division with phase-locked optical parametric oscillators [35]. David Wineland has proposed to synchronize the cyclotron motion of a single electron to a laser wave [36]. In the meantime, all these approaches have become obsolete. Since 1998, optical frequency measurements have been enormously simplified with the advent of femtosecond laser optical frequency comb synthesizers [2,3].

FEMTOSECOND LASER OPTICAL FREQUENCY COMBS

The scheme of a frequency comb synthesizer is rather simple, as illustrated in Fig. 2. At the heart is a single mode-locked femtosecond laser which maintains a soliton-like short pulse circulating inside the optical cavity. This laser can be compared to Einstein's Gedanken light clock. With each roundtrip, an attenuated copy of the light pulse escapes so that the laser emits a regular train of ultrashort pulses. To measure the unknown frequency of a laser wave, the beam and the pulse train are superimposed with a beam splitter, and a photo detector registers an interference signal. In the idealized case of a perfectly periodic pulse train, we would expect a low frequency beat note whenever the laser frequency comes close to a value where an integer number of oscillations fits in the time interval between two pulses. To give an example, if

we know that the laser emits precisely one billion pulses per second and if we can be sure that the laser wave oscillates precisely 500 000 times during the pulse repetition period, then we know that the optical frequency must be 500 000 billion cycles per second.

In the frequency domain, we can argue that the coupled longitudinal modes of the pulsed laser form an evenly spaced comb of spectral lines. A low frequency beat note is expected whenever the unknown laser frequency approaches one of these comb lines. The origin of the comb spectrum is well explained by Antony E. Siegman in his classic textbook [37]. Consider an arbitrary optical waveform circulating inside an optical cavity. During each roundtrip, an attenuated copy escapes through a partly transmitting mirror. A single copy will have a broad and more or less complicated spectrum. However, two identical copies end-to-end will produce interference fringes in the spectrum, somewhat reminiscent of Young's double slit experiment. Three copies produce a spectrum that resembles the interference pattern of a triple-slit, and an infinite series of copies produces sharp lines which can be identified with the modes of the cavity. Mathematically, an ideal periodic pulse train can be described in terms of a Fourier series, and the comb lines correspond to the elements of this series.

The separation between two modes or comb lines is just equal to the repetition frequency f_r. This remains true even if the pulses are not identical replicas but if we allow for a (reproducible) slip of the phase of the electromagnetic "carrier"-wave relative to the pulse envelope from pulse to pulse [38, 2, 3]. Such phase slips are unavoidable in a real laser because of dispersion in the cavity. The entire comb will then be shifted relative to the integer harmonics of the repetition frequency f_r by a carrier-envelope offset frequency f_{CE}, that equals the net phase slip modulo 2π per pulse interval. The frequency of a comb line with integer mode number m is then given by

$$f_m = m\, f_r + f_{CE}.$$

Such a comb acts like a ruler in frequency space that can be used to measure a large separation between two different optical frequencies in terms of the pulse repetition rate f_r. If these two frequencies are known multiples or fractions of the same laser frequency f, such a measurement reveals the optical frequency f itself. With a known repetition frequency f_r, the beat signal between a known optical frequency f and the nearest comb line reveals the previously unknown offset frequency f_{CE}. The frequency of any comb line can be calculated from the two radio-frequencies f_r and f_{CE} together with the integer mode number.

It has been surprising to most experts how far this frequency comb approach can be pushed. The frequency spectrum of a femtosecond laser oscillator can be broadened in a nonlinear optical medium to span more than an optical octave without destroying the integrity of the comb lines. In a now common implementation, the pulse train from a Kerr-lens mode-locked Ti:sapphire laser is sent through a micro structured silica fiber, with a small

solid fiber core surrounded by air-filled holes [39, 40]. The large change in refractive index at the silica-air interface permits guiding by total internal reflection even if the incoming beam is tightly focused to a high intensity. Since part of the light travels as an evanescent wave in air, an additional engineering parameter is available in such a fiber to reduce the spreading of an injected pulse due to group velocity dispersion. Inside the fiber, the pulse spectrum is broadened by self-phase modulation due to the intensity dependent refractive index, soliton splitting, shock wave formation, and other nonlinear optical processes. The emerging white light can be dispersed with a grating to form a rainbow of colors. However, this is not ordinary white light. Remarkably, the processes generating the white light can be so highly reproducible that successive pulses are still correlated in their phases and can interfere in the spectrum to form a comb of several hundred thousand sharp spectral lines.

By now, it has been confirmed in many experiments that the line spacing is very precisely equal to the repetition frequency f_r. With a comb spanning more than an octave, it is particularly simple to measure the carrier envelope offset frequency f_{CE}. One can simply select a few thousand comb lines from the red end of the spectrum and send the light pulses through a frequency doubling crystal, so that new comb lines are generated which are now displaced by twice the offset frequency f_{CE}. A collective beat note with the corresponding original comb lines near the blue end of the spectrum directly reveals the shift f_{CE}. Once this frequency can be measured, it can be controlled, for instance by adjusting the dispersion in the laser cavity or simply by changing the pump power. One can even set f_{CE} to zero so that the frequencies of the comb lines become precise integer multiples of the laser repetition frequency f_r.

So far, we have treated all light waves as classical electromagnetic waves. The quantum optical aspects of frequency combs, i.e. expected correlations in the noise due to photons and their entanglement, have not yet been explored. Such studies may lead to a rich new field of research.

A laser frequency comb provides a direct link between optical frequencies and microwave frequencies. This link can be used in either direction. We can measure or control the repetition frequency f_r with a cesium atomic clock to synthesize several hundred thousand sharp optical reference frequencies which are precisely known in terms of the primary standard of time. Any unknown frequency can then be determined by first making a wavelength measurement with a conventional wave meter that is sufficiently accurate to determine the integer order number m of the nearest comb line. The precise distance from this reference line is then measured by feeding a beat signal to a microwave counter. In the reverse direction, we can start with a sharp optical reference line in some cold trapped ion, cold atoms, or slow molecules, and lock a nearby comb line to this optical reference. All the other comb line frequencies are then rational multiples of the optical reference frequency, and the repetition frequency becomes a precisely known fraction.

Frequency comb synthesizers act as if we had several hundred thousand ultra-stable and precisely tuned lasers operating at once. With the help of

nonlinear sum and difference frequency generation, they can precisely measure any frequency from radio waves to the near ultraviolet. They provide the long-missing clockwork for optical atomic clocks. They can even generate microwaves with extreme phase stability [41]. As sources of phase-stabilized femtosecond pulses they have even given us a key to the intriguing field of attosecond science [42]. With their electronic servo controls, frequency-comb synthesizers can be relatively simple, robust, and increasingly user-friendly devices.

THIS IS A SIMPLE IDEA! WHAT TOOK SO LONG?

In hindsight, the ideas behind the optical frequency comb look rather simple and almost obvious. Why did all the experts, including our own laboratory, struggle for so long with much more cumbersome harmonic laser frequency chains?

The main reason may be that nobody believed seriously that such an approach could actually work. There were good arguments why it should be impossible to bridge the gap between radio frequencies and optical frequencies in a single step. The phase noise of even the best quartz oscillator is so large that any comb structure would be completely washed out if one could somehow multiply its frequency up into the visible region. In a harmonic frequency chain, the intermediate transfer oscillators act as phase noise filters and electromagnetic "flywheels" to overcome this "coherence collapse" [43].

Another reason may be that two separate scientific communities have evolved since the early days of laser science. People interested in precise high resolution spectroscopy used their ingenuity to perfect the frequency stability of continuous wave lasers. On the other side, there were people who invented clever techniques to produce ever shorter pulses with mode-locked lasers. They applied their spectrally broad light flashes to the study of ultrafast phenomena in semiconductors, liquids, or in chemical reaction dynamics or to generate ever higher peak intensities for experiments in plasma physics. These two communities went to their own separate conferences, and neither side felt a strong need to keep track of the other frontier.

For our own work, I cannot hide behind this latter excuse. I knew since the early experiments with multi-mode helium-neon lasers [44] that the longitudinal modes of a laser are well defined and their phases can be coupled so as to produce a short light pulse circulating inside the cavity [45, 46]. Much shorter pulses were produced some years later with broadband dye lasers by locking their axial modes with the help of a saturable absorber or by synchronous pumping with a modulated argon laser [47]. At Stanford in the mid-seventies, I became intrigued by the idea of high resolution spectroscopy of atomic resonance lines by Ramsey-like excitation with a coherent train of multiple light pulses [48]. Resonant excitation with separated light pulses has also been explored at the time by Michael Salour at MIT [49] and by Veniamin Chebotaev at Novosibirsk [50]. After initial encouraging experiments with a dye laser pulse injected into a passive cavity [48], our group at Stanford

with graduate student Jim Eckstein and visiting Lindemann Fellow Allister Ferguson demonstrated that a synchronously pumped mode-locked picosecond dye laser could produce a stable phase coherent pulse train which we used for Doppler-free two-photon excitation of atomic sodium [51]. The comb lines served as a frequency ruler to measure some atomic fine structure intervals. To improve the precision, we replaced the original radio frequency driver for the modulator of our argon pump laser with a high quality frequency synthesizer. Much to our delight, the performance of the dye laser improved so much that we were the first to generate sub-picosecond pulses directly from a synchronously pumped dye laser [52]. At that time we should have learned an important lesson: what is good for frequency stability is also good for the generation of ultrashort light pulses! We later explored Doppler-free polarization spectroscopy with our frequency comb [53], as well as two-photon spectroscopy with the frequency comb of an FM mode-locked laser where the phases of the modes adjust so that the intensity remains constant but the frequency is sweeping back and forth periodically [54].

During the Stanford experiments, we were painfully aware that we could not know the absolute positions of our comb lines because the dispersion inside the laser resonator would lead to unknown phase slips of the carrier wave relative to the pulse envelope. Such phase slips shift the entire comb spectrum by an unknown amount f_{CE}, as worked out in considerable detail in the 1978 Stanford Ph.D. thesis of Jim Eckstein [38]. With a comb spectrum spanning only 800 GHz, we had no means to observe and measure the offset frequency f_{CE}. Therefore, we did not know how to measure absolute optical frequencies with our laser frequency combs in the late seventies.

The idea of somehow generating much broader frequency combs surfaced again in my mind after I had moved back to Germany in 1986. In 1990, I published a proposal for a synthesizer of sub-femtosecond pulses that would superimpose the frequencies from separate phase-locked cw laser oscillators to form a very wide comb [55]. In the early 1990s, the technology of ultrafast lasers advanced dramatically with the discovery of Kerr-lens mode locking by Wilson Sibbett at the University of St. Andrews [56]. Soon, commercial Ti:sapphire femtosecond lasers became available, that made the generation of ultrashort light pulses much easier. Intrigued by these new sources I discussed with Peter Lambropoulos at Garching the possibility of finding some highly nonlinear effect, such as above-threshold-ionization (ATI) , that would depend on the phase of the electric field relative to the pulse envelope and could reveal the offset frequency f_{CE} of the laser comb lines. Calculations soon showed that such effects would be observable only for pulses lasting at most a few optical cycles [57]. Today, such sources have become available, and the phase dependence of ATI has since been demonstrated by Gerhard Paulus and Herbert Walther [58]. In 1994, I also discussed the problem of the carrier-envelope phase slips with Ferenc Krausz at the Technical University of Vienna. His group was the first to observe such pulse-to-pulse phase slips in an interferometric correlation experiment in 1996 [59].

I remember a trade show in 1994, when I was captivated by an exhibit of a

(Coherent Mira) mode-locked Ti:sapphire femtosecond laser with regenerative amplifier. The laser beam was focused into a glass slide to produce a white light continuum which a prism dispersed into a rainbow of colors. Such white light pulses are produced by a combination of self-focusing, self phase modulation, and other nonlinear processes, and they have been used for a long time in ultrafast pump-probe experiments [60]. A striking feature was the laser-like speckle pattern in the rainbow colors which indicated a high degree of spatial coherence. It occurred to me that such a system might produce an octave-spanning frequency comb if the phases of successive pulses were sufficiently correlated. Such a wide comb could then be used as a ruler to measure the large interval between a laser frequency and its second harmonic, which must be equal to the laser frequency itself. Even though the pulse repetition frequency of a few hundred kHz remained inconveniently low for frequency comb experiments, I felt sufficiently intrigued to acquire such a system for our frequency metrology laboratory at Garching in 1994. In the back of my mind I had the hope that it might somehow be possible to produce white light directly with the pulses from the laser oscillator, without the regenerative amplifier with its much lower pulse rate, by sending the pulse train into a small waveguide made from some highly nonlinear optical material, so that it would not be necessary to reach the threshold power for self-focusing.

We did not pursue the femtosecond laser approach seriously right away, because we had come quite far in perfecting our alternative scheme of optical interval division. An accurate measurement of the 1S-2S frequency seemed almost within reach. We also felt that we would need an independent tool to verify any measurement with a femtosecond laser frequency comb, since the frequency metrology community would otherwise distrust our results. The hydrogen measurements were finally completed in 1997 [29, 30].

In February 1997, I visited the European Laboratory for Nonlinear Spectroscopy, LENS, in Florence, Italy. There, Marco Bellini was working with an amplified Ti:sapphire femtosecond laser producing pulses of 1 mJ energy at a rate of 1 kHz. As is common in many ultrafast laboratories, he produced a white light continuum for pump-probe experiments by focusing part of the laser beam into a thin plate of CaF_2. I asked what would happen if we split the laser beam in two parts and focus these beams at two spatially separate spots. Would the two white light pulses interfere?

In an earlier joint experiment at the Lund Laser Center, we had investigated the same question for the generation of high harmonic radiation in a gas jet [61]. From that time, Marco Bellini still had a Michelson interferometer on his shelve that we could quickly place into the laser beam, somewhat misaligned so that two beams would escape in two slightly different directions. By adjusting the length of one arm we could make sure that the two focused pulses arrived on the CaF_2 plate at precisely the same time. I felt electrified when we observed stable interference fringes of high contrast for all the colors that I could record with my handheld camcorder electronic notebook, as shown in Fig. 3 [62]. The white light pulses had to be phase-locked to the

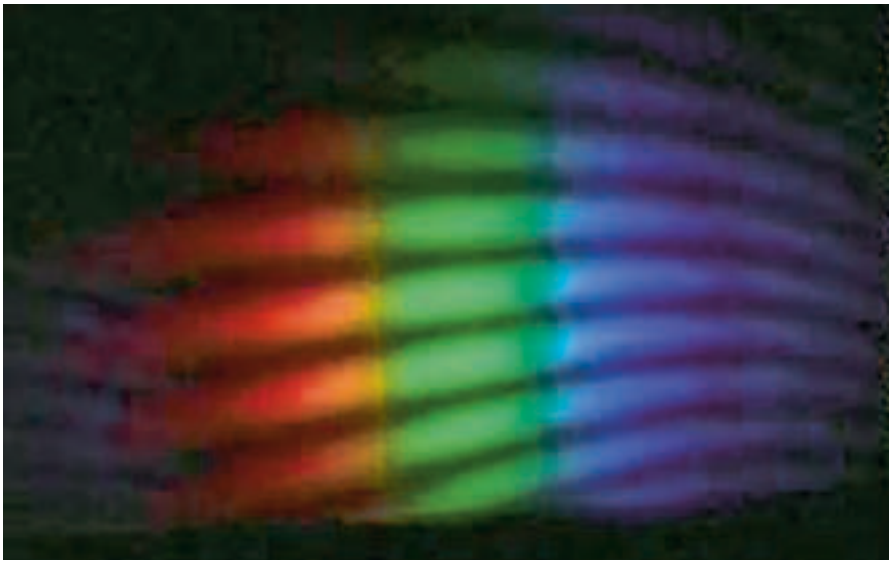

Figure 3. Interference fringes between two white light pulses [Ref. 62].

driving laser field! No matter how complicated the process of white light continuum generation might be, the process was reproducible. If such pulses were separated in time rather than in space, they would interfere in the spectrum to produce a very broad frequency comb.

By March 30, 1997, I had written a confidential 6-page proposal for a universal optical frequency comb synthesizer "…*which produces a wide comb of absolutely known equidistant marker frequencies throughout the infrared, visible, and ultraviolet spectral range. To this end, a white light continuum with a pulse repetition rate* f_r *is produced by focusing the output of a mode-locked femtosecond laser into an optical fiber or bulk medium with a third order nonlinear susceptibility. The rate of phase slippage of the laser carrier relative to the pulse envelope,* f_{CE}*, is monitored by observing a beat signal between the white light continuum and the second harmonic of the laser.*" The envisioned self-referencing scheme could find the carrier-envelope offset frequency f_{CE} without any auxiliary laser. I asked Thomas Udem and Martin Weitz in our laboratory to witness and sign every page on April 4, 1997, since this might become important for later patent applications.

At Garching, we soon started a serious experimental effort towards optical frequency measurements with femtosecond laser frequency combs. Even if we did not yet know how to broaden the spectrum of our Mira laser oscillator to more than an optical octave, we could always follow the 1988 proposal [27] and measure the frequency of the dye laser in our hydrogen spectrometer by implementing a short chain of two or three interval divider stages, using small semiconductor lasers, to arrive at a frequency gap that could be bridged with our laser comb. Propelled by such visions, Thomas Udem and Jörg Reichert investigated the frequency comb spectrum of the Mira femtosecond

laser. They were later joined by Ronald Holzwarth. By that time, hundreds of such lasers were in use in laboratories around the world, but they were mostly used to study ultrafast phenomena. Nobody had ever looked for any comb lines, as far as we could tell. With a repetition frequency of 76.5 MHz, the comb spectrum of our femtosecond laser was so densely spaced that no spectrometer in our laboratory could resolve the comb lines. Therefore, we resorted to heterodyne detection, employing a cw diode laser as a local oscillator. The diode laser beam and the pulse train were superimposed with a beam splitter, and a beat signal was detected with an avalanche photodiode after some spectral filtering. After paying attention to the mechanical stability of the femtosecond laser, we could observe stable comb lines. Next, we investigated the spacing of these lines. We phase-locked two diode lasers to two arbitrarily chosen comb lines and used an optical interval divider stage to produce a new frequency precisely at the center. A beat note with the nearest comb line confirmed, much to our delight, that the comb lines were perfectly evenly spaced, way out into the wings of the emission spectrum, within a few parts in 10^{17} [63].

It was now certain that the frequency comb of such a mode-locked femtosecond laser did not suffer from "coherence collapse" and could serve as a ruler in frequency space to measure large optical frequency intervals. In a first demonstration of an optical frequency measurement with a femtosecond laser comb, we determined the frequency interval between the cesium D1 resonance line and the fourth harmonic of a transportable CH_4-stabilized 3.39 μm He-Ne-laser, which had been calibrated with a harmonic laser frequency chain at the PTB Braunschweig [64]. The optical cesium frequency was needed for a determination of the fine structure constant α from the atomic recoil energy as measured by atom interferometry in the group of Steve Chu at Stanford. Soon these experiments found a considerable resonance in newspapers and journals. They demonstrated to the optical frequency metrology community that femtosecond laser frequency combs could be powerful tools to measure the frequency of light.

We next turned to the more ambitious goal of measuring the absolute optical frequency of our 486 nm dye laser in the hydrogen 1S-2S spectrometer. At that time, we had broadened the frequency comb of our mode-locked laser oscillator by self-phase-modulation in a short length of ordinary optical fiber to span 60 or 70 THz. Rather than building a few new interval divider stages, we recognized a more expedient approach for a first proof-of-principle experiment. Our hydrogen spectrometer [29] required only minor modifications to produce two different fractional subharmonics, 4/7 and 1/2, of the dye laser frequency, which we could bridge with our femtosecond laser comb [65, 66]. The CH_4-stabilized helium neon laser served now as part of an interval divider and no longer as an intermediate reference standard. The primary reference for our first absolute frequency measurements was a commercial HP cesium atomic beam clock which we used to determine the pulse repetition rate f_r and the carrier-envelope offset frequency f_{CE} [65]. Once the optical frequency gap was measured, we knew the absolute frequency of the

dye laser as well as the absolute frequencies of all the comb lines. To control the position of the comb lines, we learned how to change the frequency f_{CE} of our Mira Ti:sapphire laser by tilting an end mirror where the spectrum is slightly dispersed by a prism pair inside the cavity. In this way, we were the first to generate femtosecond laser pulses with controlled slips of the carrier-envelope phase.

In October 1998 we proudly showed our experiment to Norman Ramsey, who had come to the Max-Planck-Institute as a member of the Scientific Advisory Board. For the first time, we could compare the hydrogen 1S-2S frequency directly with a cesium atomic clock in our own laboratory, without involving any large harmonic laser frequency chain. Later in the same year, we demonstrated our experiment to John Hall who had come to Munich to attend a meeting commemorating our mutual friend Veniamin Chebotaev. John soon became an ardent evangelist for "this goofy technique, that makes everything obsolete that we have worked on for so long." He started to assemble a powerful professional team in Boulder to advance research on femtosecond laser frequency combs, and he persuaded his colleague Steve Cundiff at JILA, an expert on femtosecond lasers from Bell Laboratories, to visit our laboratory in the spring of 1999. An increasingly heated competition did much to accelerate the development of the new tools in the coming months and to ignite a firework of novel applications in the years to follow [3].

Until the summer of 1999, we had felt as if the new femto-comb playing ground belonged just to us. Thomas Udem had reported on our experiments at a conference in Perth, Australia, in late 1998 [43]. But we had delayed publications such as a first article on "measuring the frequency of light with mode-locked lasers" [67] to appear after the filing of our first patent application in March 1999 because, according to German law, an invention can no longer be patented once it has been published.

In June 1999, we could directly compare the hydrogen frequency with a highly accurate transportable cesium fountain clock (PHARAO), built at the LPTF (now BNM SYRTE) in Paris [66]. This measurement yielded a new value of the hydrogen 1S-2S frequency accurate to 1.8 parts in 10^{14}, surpassing all earlier optical frequency measurements by more than an order of magnitude. By now, the compelling advantages of laser frequency combs had been clearly demonstrated. A number of different possible approaches for carrier-envelope offset phase control were soon proposed [68].

As a next step in our own work, we wanted to drastically simplify our setup. Around that time, a new tool had appeared on the horizon that made it likely that we would no longer need any optical interval dividers. At the CLEO conference at Baltimore, MD, held in May 1999, researchers from Bell Laboratories had reported on a novel micro-structured "rainbow fiber" that could broaden the spectrum of pulses of a Ti:sapphire femtosecond laser oscillator without further amplification to a rainbow of colors [40]. After the white light interference experiments in Florence [62], I felt rather confident that this magic fiber would preserve the phase coherence of successive pulses and produce comb lines with a desirable large frequency spacing.

In June 1999, John Hall came to Germany to participate in the annual retreat of our research group at the Ringberg Castle near the Tegernsee south of Munich. Together, we phoned many of our old friends at Bell Laboratories, to try and obtain a sample of the magic fiber. We were hoping that we could demonstrate an octave-spanning frequency comb while John Hall was still in Germany. Unfortunately, this plan was foiled by the lawyers at Lucent Technologies who did not allow the fiber to leave Bell Laboratories. Ronald Holzwarth traveled to Bell Laboratories at Holmdel, NJ, during his 1999 summer vacation, but he had to leave without a piece of the fiber. John Hall's team in Boulder experienced similar difficulties at first, but in October 1999, they could demonstrate the first octave-spanning self-referencing laser frequency comb after finally securing some of the holey fiber [69, 70]. At Garching we realized a similar comb system a few weeks later [71], after we had received some "photonic crystal fiber" from the group of Philip Russell at the University of Bath in the UK. We had found out too late that these British researchers had actually pioneered micro-structured silica fibers some years earlier [39]. Both laboratories submitted their first short publications on octave-spanning frequency combs on the same day (Nov. 12, 1999) to the CLEO/QELS 2000 conference in San Francisco.

Similar to the Boulder experiments, we used a commercial small Ti:sapphire ring laser for our first octave-spanning frequency comb, producing pulses of about 25 fs duration at a repetition frequency of 625 MHz. Launching about 170 mW into a 30 cm length of photonic crystal fiber, we immediately produced a frequency comb spanning more than an octave. The spectrum showed a complicated structure, with valleys and peaks, but it offered useable comb lines everywhere. Together with a nonlinear interferometer for control of the offset frequency f_{CE} , the entire optical setup did easily fit on a single breadboard. While the traditional harmonic frequency chains with their factory halls full of lasers could measure just one single optical frequency, our new system was ready to measure any frequency throughout the visible and near infrared.

Since then, Ti:sapphire femtosecond lasers have been developed that produce an octave-spanning spectrum directly from the oscillator, without any need for external spectral broadening [72]. Octave spanning combs can also be generated with erbium-doped fiber lasers [73], pumped by very reliable and robust laser diodes developed for telecommunications. As turn-key instruments, such fiber comb generators can run for months without human attention.

In a first stringent test, Ronald Holzwarth has compared an octave spanning frequency comb synthesizer with the more complex frequency synthesizer used in the 1999 hydrogen frequency measurement [71]. By starting with a common 10 MHz radiofrequency reference and comparing comb lines near 350 THz, he could verify agreement within a few parts in 10^{16}, limited by Doppler shifts due to air pressure changes or thermal expansion of the optical tables. In 2002, a group at the PTB in Braunschweig demonstrated how a femtosecond laser frequency comb generator can be used as a transfer oscil-

lator to precisely measure optical frequency ratios [74]. As a test case, they measured the frequency ratio between the second harmonic of a Nd:YAG laser and the fundamental frequency, verifying the expected value of 2 with an uncertainty of 7 parts in 10^{19}. More recently, Marcus Zimmermann in our laboratory has pushed a related experiment to an uncertainty of 6 parts in 10^{21} [75]. In 2004, researchers in Boulder compared four different frequency combs from different laboratories, finding agreement between neighboring comb lines at an uncertainty level of 10^{-19} [76]. So far, no systematic error has been identified which would limit the potential accuracy of future precision spectroscopy or optical atomic clocks.

NEW FREQUENCY MEASUREMENT OF HYDROGEN 1S-2S IN 2003: ARE THE FUNDAMENTAL CONSTANTS CONSTANT?

In February 2003, we used an octave spanning comb synthesizer in a new measurement of the hydrogen 1S-2S transition frequency [21]. Marc Fischer and Nikolai Kolachevsky had implemented many improvements in the hydrogen spectrometer. Light from the dye laser was sent through a fiber into the frequency metrology laboratory, where an octave-spanning Ti:sapphire femtosecond laser frequency comb synthesizer was used to compare the optical frequency to the radio frequency of the Paris PHARAO atomic fountain clock, which had again been brought to Garching.

With such immediate absolute frequency calibration, the hydrogen spectroscopy could be performed with much confidence. Compared to the 1999 measurements, the statistical error of the data recorded on a given day was much reduced. Nonetheless, the day-to-day fluctuations of our measurements remained of similar magnitude as before. They indicate some uncontrolled systematic errors. After careful further experiments probing for possible causes of systematic line shifts and after a thorough statistical analysis of all recorded data, we believe that the fluctuations are caused by some residual small first order Doppler shifts. Such shifts are expected if the two counter-propagating wave fronts of the exciting 243 nm radiation do not match perfectly. A mismatch may be caused by imperfect mode-matching of the frequency-doubled dye laser beam that enters the build-up cavity inside the atomic beam apparatus through a 2% input coupling mirror. Another cause may be the accumulation of frozen molecular hydrogen on the walls of the cold copper nozzle for the hydrogen atomic beam, which can grow until it distorts the optical wave fronts by vignetting and diffraction. A second cause of systematic line shifts in two-photon spectroscopy with frequency-doubled laser light may be unwanted correlations between amplitude noise and phase noise that could be caused by imperfect servo-locks of the enhancement cavities. For the future, we are preparing an ultraviolet build-up cavity of higher finesse, and we are working towards an all-solid-state laser source with a line width of only a few Hz.

From the 2003 measurement, we find a frequency of 2 466 061 102 474 851 ± 34 Hz for the F=1 to F'=1 hyperfine component of the hydrogen 1S-2S frequency, with a relative uncertainty of 1.4 parts in 10^{14}. The new results agrees

within the error limits with the 1999 measurement of 2 466 061 102 474 880 ± 46 Hz. A difference of 29 ± 57 Hz in 44 months corresponds to a relative drift of the 1S-2S transition frequency of $(3.2 \pm 6.3) \times 10^{-15}$ per year, i.e. it is compatible with zero drift.

This experiment has attracted some attention because it can be considered as a test for a possible slow variation of the electromagnetic fine structure constant α. During the 2003 experiment, theorist Harald Fritzsch called frequently to get some preliminary results, because he had predicted an observable drift of the microwave frequency of the cesium clock relative to the hydrogen frequency [77]. Starting point for his arguments were astronomical observations of spectral lines in the light of distant quasars performed at the Keck Observatory [78]. Differential red shifts seemed to suggest that the electromagnetic fine structure constant α in the early universe was somewhat smaller than today. Making the simplest assumption of a linear drift, the data would indicate a drift of $(6.4 \pm 1.35) \times 10^{-16}$ per year, too small to be observable in our laboratory experiment. However, Fritzsch had argued with ideas from grand unification and quantum chromodynamics that α cannot change simply by itself. If all known forces are to remain unified at very high energies, other coupling constants must change as well. As a result, the masses and magnetic moments of hadrons (in units of the Bohr magneton) should change relative to those of the electron. Fritzsch pointed out a possible magnifying effect, that could change the hyperfine transition frequency of the cesium atomic clock about 20 times faster than the optical hydrogen frequency. So far, we have not found any evidence for such a drift. There are also other more recent observations of quasar spectra that do not support the evidence for a changing fine structure constant [79].

Regardless of such speculations, we have to admit that the hydrogen measurements of 1999 and 2003 do not strictly rule out a changing fine structure constant α. It is conceivable that the magnetic moment of the cesium nucleus also changes at just the right rate to give a null result in our experiment. Fortunately, such measurements are not limited to hydrogen. One can also measure transition frequencies in heavier atoms with stronger relativistic effects that respond in a different way to changes in α. One such candidate is the clock transition in a single cold Hg^+ ion, that has been compared by Jim Bergquist and his team at Boulder with a cesium atomic clock in 2000 and 2002, also using a laser frequency comb [80]. In addition, Eckhard Peik and his team at the PTB have made two separate measurements of the clock transition in a single Yb^+ ion [81]. Together, these laboratory experiments give now some upper limits for the possible rates of change of the fine structure constant α and the cesium nuclear magnetic moment μ_{Cs} on the order of $(-0.3 \pm 2.0) \times 10^{-15}$ per year and $(2.4 \pm 6.8) \times 10^{-15}$ per year, respectively.

Within the next few years we can expect much more stringent experimental limits on possible variations of physical constants from such laboratory experiments. If we found such changes it would not have any consequences for our everyday lives, but it would give reason for fascinating speculations about the nature of the universe.

Sensitive limits for the variations of fundamental constants will be established in the comparison of different types of optical atomic clocks that are now being developed by strong professional teams in many industrialized countries. The perfection of optical frequency standards has been progressing at a much more rapid pace than that of microwave cesium clocks [82]. With the femtosecond laser comb now available as a perfect clockwork mechanism, the efforts must concentrate on ever more perfect laser frequency stabilization and on the management of systematic line shifts in precision spectroscopy of narrow optical resonances that serve as the "pendulum" of optical clocks. Much progress has already been made in experiments with cold trapped ions, notably Hg^+, Yb^+, In^+, and Sr^+. Cold neutral atoms such as H, Ca, or Sr are also appealing candidates because many atoms can be observed simultaneously without disturbing Coulomb repulsion, improving the signal-to-noise ratio and the speed with which a resonance frequency can be established. A particularly promising approach has been proposed by Hidetoshi Katori [83]. In his neutral atom clock, many cold neutral Sr atoms are captured in the microscopic dipole force potential wells of an optical lattice. Light shifts are minimized by choosing a proper "magic" wavelength of the lattice field. In 2005, the accuracy of the best optical frequency standards has become comparable to that of the best cesium fountain clocks. But even if they are not yet more accurate, optical frequency standards offer one important advantage already. It takes hours or days to compare two cesium clocks to a part in 10^{15}. Two optical frequencies can be compared to this level within just seconds.

It is interesting to look at the historical evolution of the accuracy of clocks. The clocks in medieval church towers were only good to about 20 minutes per day. In the 18^{th} century, the nautical clock H4 of the legendary watchmaker John Harrison reached an accuracy of some 100 msec per day. The best primary cesium fountain clocks of today can be accurate to within 100 psec per day. Some experts hope that optical atomic clocks will reach a hundred or thousand fold higher accuracy within the next decade.

Better atomic clocks will be enabling tools for many scientific and technical applications, so that this pursuit will be worthwhile even if we do not discover any changes of fundamental constants. They can extend the frontiers of precision spectroscopy and of time and frequency metrology. They will make it possible to precisely synchronize clocks over large distances. In astronomy, such synchronized clocks may allow an extension of large baseline interferometry to infrared and optical wavelengths. Better clocks can improve the performance of satellite navigation systems and the tracking of probes in deep space. Accurate clocks are also needed to synchronize optical telecommunication networks. In fundamental physics, more accurate clocks will permit more stringent tests of special and general relativity, as well as other fundamental laws.

So far, we have not discovered any fundamental limits for the potential accuracy of future clocks. It should even be possible to extend frequency comb techniques into the extreme ultraviolet and soft X-ray spectral regions, so that we would be able to slice time into still much finer intervals. High harmonic radiation that is generated when an intense femtosecond laser pulse is focused into a gas jet can be harnessed to produce coherent pulse trains at such wavelengths.

Since the pioneering work of Charlie Rhodes [84] and Anne L'Huiller [85] in the late 1980s, high harmonic generation has been studied in many laboratories. In a simple model first proposed by Paul Corkum [86], gas atoms are field ionized and the electrons are accelerated by the strong laser field until the light field reverses direction. Dependent on the time of escape, such electrons can return to the ion core with substantial kinetic energy which they can radiate in the form of energetic photons, with one burst emitted during each half cycle of the driving laser wave.

The mutual phase coherence of short pulses was much on my mind when I visited the high harmonic experiments of Anne L'Huiller and Claes Göran Wahlström at the Lund Laser Center in 1995. I wondered if two successive high harmonic pulses would be mutually phase coherent. As a test I proposed to split the driving laser beam into two parts that could be focused into the gas jet at separate spots, and to look for interference fringes in the high harmonic radiation, similar to the later white light experiments at Florence [62]. At first, there seemed to be good reasons why this was not to be expected, because the phase of the harmonic radiation should depend strongly on the varying intensity of the driving laser pulse. But after I left, graduate student Raoul Zerne tried the experiment and observed some fleeting interference fringes. With much excitement, we scheduled some serious joint experiments at Lund. Marco Bellini from LENS agreed to participate and to construct a stable Michelson interferometer so that the timing of the two laser pulses could be finely adjusted. We soon observed clean interference fringes of high contrast up to the 15^{th} harmonic and beyond [61]. In a subsequent experiment we even discovered a regime where the harmonic beam was surrounded by a divergent halo beam of very short coherence length. This behavior could be explained in terms of two different electron trajectories that can contribute to a given harmonic photon energy [87].

These results demonstrate that high harmonic pulses can be mutually phase coherent so that a regular train of such pulses could form a frequency comb in the extreme ultraviolet. However, the necessary peak intensities of the order of 10^{14} W/cm^2 could only be produced with amplified femtosecond laser systems of low repetition frequency. Very recently, Christoph Gohle and Thomas Udem at Garching have succeeded in producing high harmonic radiation down to wavelengths of 60 nm at a repetition frequency of 112 MHz [88]. To this end, they stacked the pulses from a mode-locked Ti:sapphire laser oscillator in a dispersion-compensated passive build-up cavity and

placed a xenon gas jet at an intra-cavity focus. The high harmonic radiation is coupled out by external reflection from a thin sapphire Brewster plate, that has a refractive index smaller than 1 in the extreme ultraviolet. Similar experiments have also been reported by Jun Ye in Boulder [89].

In a future ambitious project, we plan to apply frequency combs in the XUV directly to precision spectroscopy of sharp resonances in laser-cooled trapped ions. The hydrogen-like helium ion with a 1S-2S two-photon transition near 60 nm is a particularly interesting candidate. In one envisioned scenario, helium ions will be sympathetically cooled by laser-cooled magnesium ions in the same trap, and the signal might be detected via the production of doubly charged helium ions due to photo ionization.

CONCLUSIONS

Spectroscopy of the simple hydrogen atom has sparked off the cross fertilization of two seemingly unrelated frontiers, precise optical spectroscopy and the study of ultrafast phenomena. Femtosecond frequency combs are revolutionizing precision measurements of time and frequency. Future optical atomic clocks will find important applications in many areas of science and technology. Ultraprecise optical spectroscopy can be harnessed for new tests of fundamental physics laws. However, many other spectroscopic applications of laser frequency combs can be envisioned, such as massively parallel ultra-sensitive cavity ring-down spectroscopy [90] or broadband spectral interferometry. At the same time, frequency comb techniques are also offering powerful new tools for ultrafast physics. By controlling the phase of the electric field of intense light pulses lasting for only a few cycles, they make it possible to study ultrafast electronic processes in light matter interactions, such as the production of single sub-femtosecond pulses of soft X-rays in high harmonic generation [42]. Only the future can show what we will discover with such exquisite new instruments.

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